Technical Program Review

Nuclear Criticality Safety Program

March 18 – 19, 2015 • LLNL, Livermore, CA, USA

Development of the Thermal Neutron Scattering Law Of Polymethyl Methacrylate (Lucite)

Ayman I. Hawari

Nuclear Reactor Program
Department of Nuclear Engineering
North Carolina State University
Raleigh, North Carolina, USA

Acknowledgement

- The many graduate students and postdocs at North Carolina State University

- Current funding by the NCSP program and past funding by DOE NE for through NERI and NEUP programs

Vision

- Establish a predictive approach for generating the needed data (cross sections) to describe the energy exchange of thermal neutrons in matter

- Various applications:
  - Nuclear criticality safety
  - Nuclear reactor design
  - Neutron beam spectral shaping (i.e., filtering)
  - Neutron source (cold, ultracold, etc.) characterization

$$\left.\frac{d^2\sigma}{d\Omega dE}\right|_{inelastic} = \frac{\sigma}{2k_BT}\sqrt{\frac{E'}{E}}e^{-\frac{\beta}{2}}S(\alpha,\beta)$$

$\beta = \dfrac{E-E'}{k_BT}$ Energy transfer

$\alpha = \dfrac{(E+E'-2\sqrt{EE'}\cos\theta)}{k_BT}$ Momentum transfer

The scattering law is the Fourier transform of a correlation function

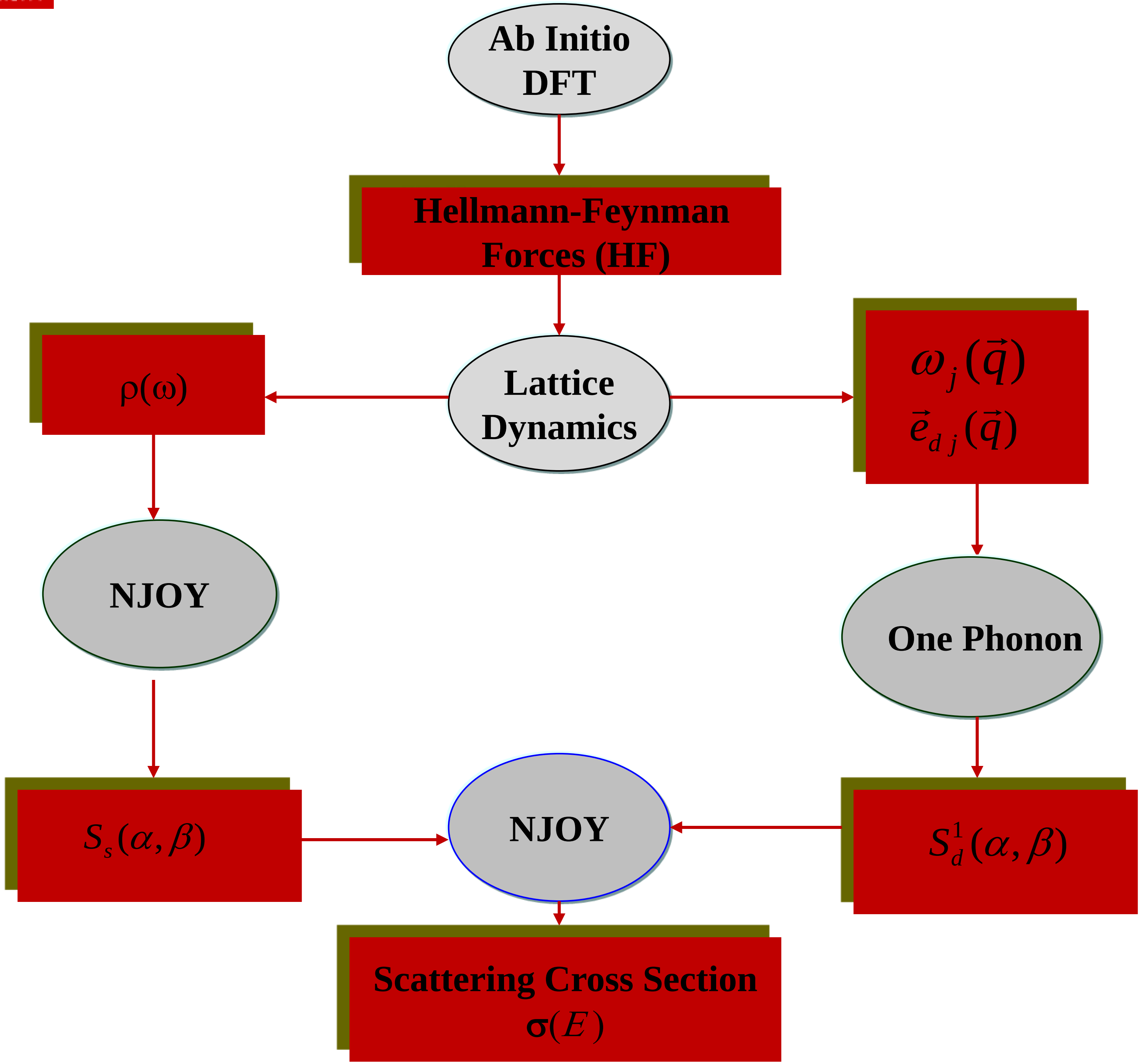
$$S(\alpha,\beta) = \frac{1}{2\pi}\int_{-\infty}^{\infty} e^{-i\beta t}e^{-\gamma(t)}dt$$

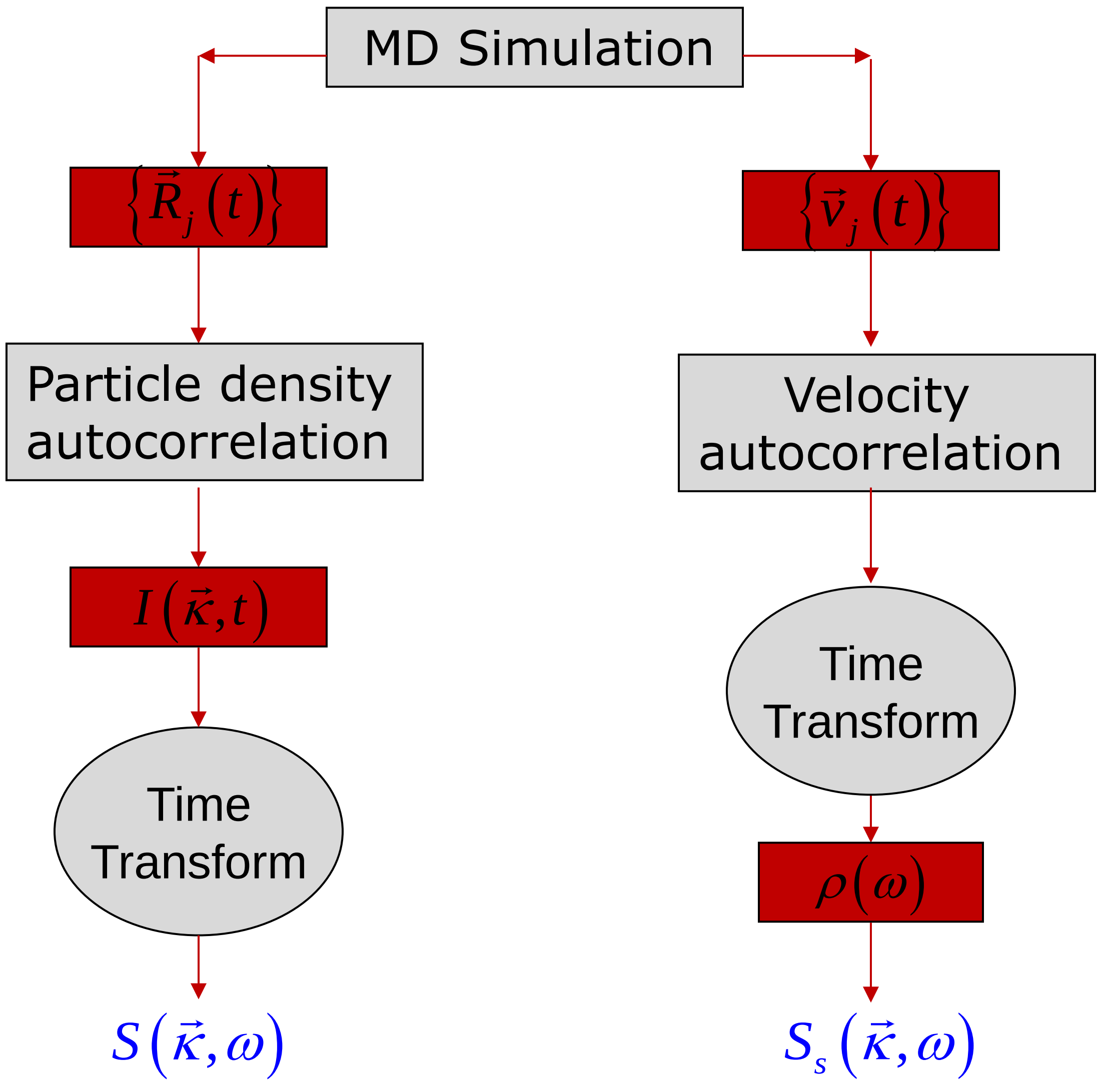
$$\gamma(t) = \frac{\alpha}{2}\int_{-\infty}^{\infty}\frac{\rho(\beta)}{\beta\sinh(\beta/2)}\left[1-e^{-i\beta t}\right]e^{\beta/2}d\beta$$

$\rho(\beta)$ – density of states (e.g., phonon frequency distribution)

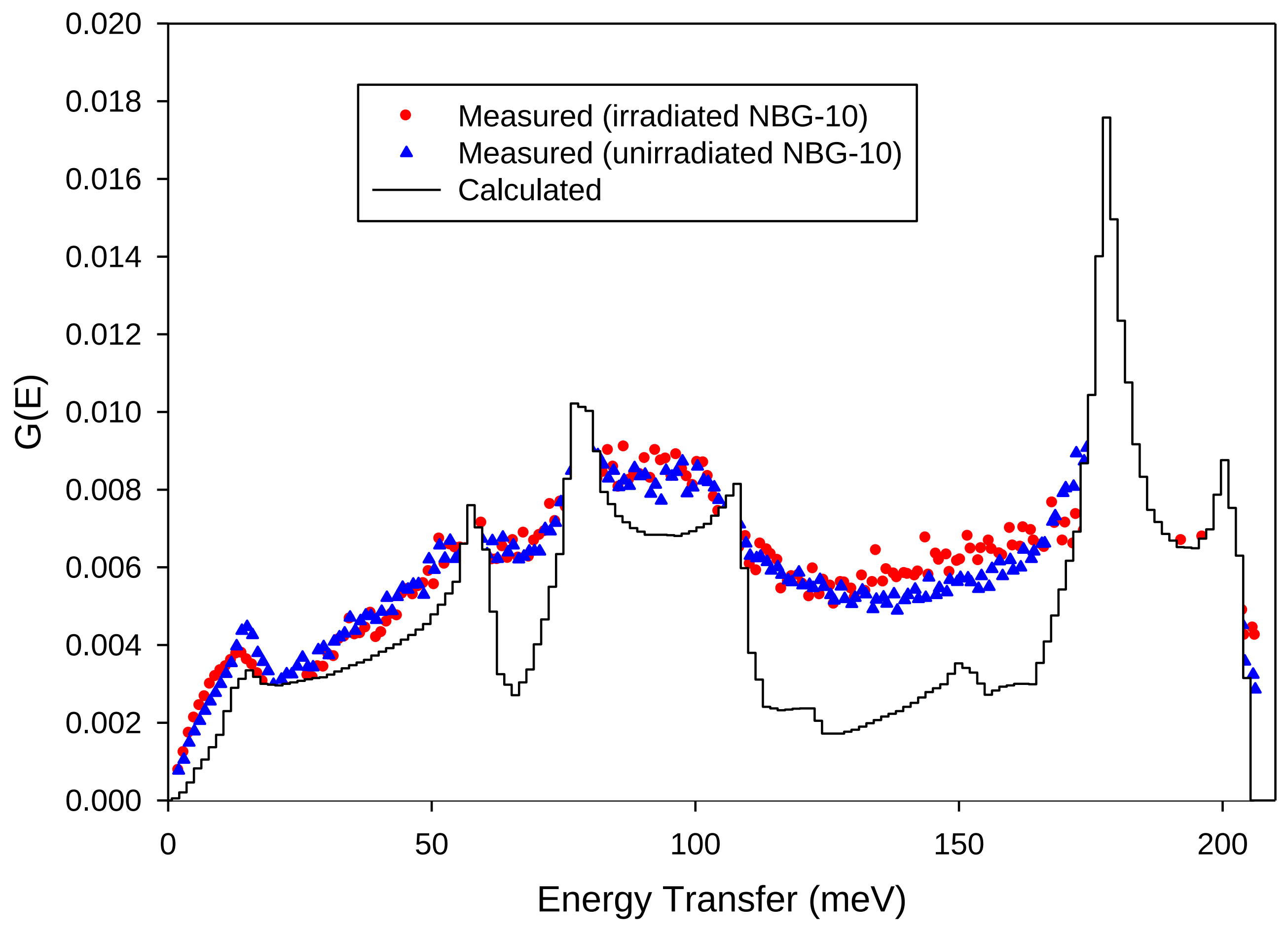
Methods

- Several approaches can be used to extract the atomic density of states, scattering law and eventually the cross sections
  - Empirical atomic force analysis combined with dynamical matrix calculations
    - Basis of current ENDF/B libraries
  - Ab initio Quantum (DFT) methods combined with dynamical matrix calculations
  - Classical Molecular Dynamics (MD) methods combined with correlation function analysis

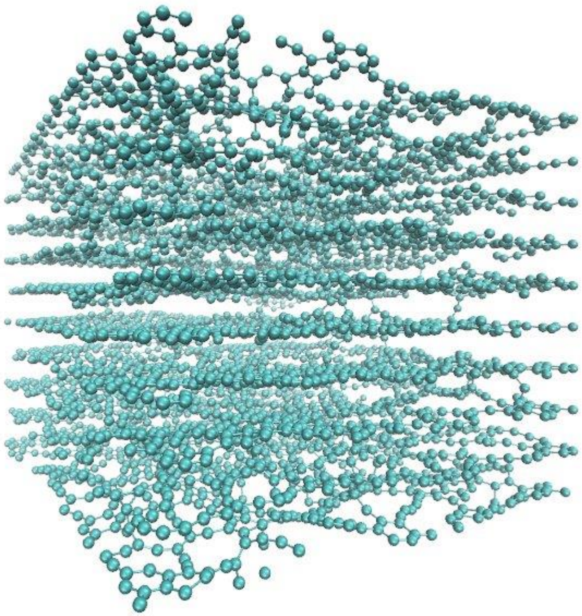

Ab Initio DFT
Hellmann-Feynman Forces (HF)
Lattice Dynamics
$\rho(\omega)$
$\omega_j(\vec{q})$
$\vec{e}_{dj}(\vec{q})$
NJOY
One Phonon
$S_s(\alpha,\beta)$
NJOY
$S_d^1(\alpha,\beta)$
Scattering Cross Section
$\sigma(E)$

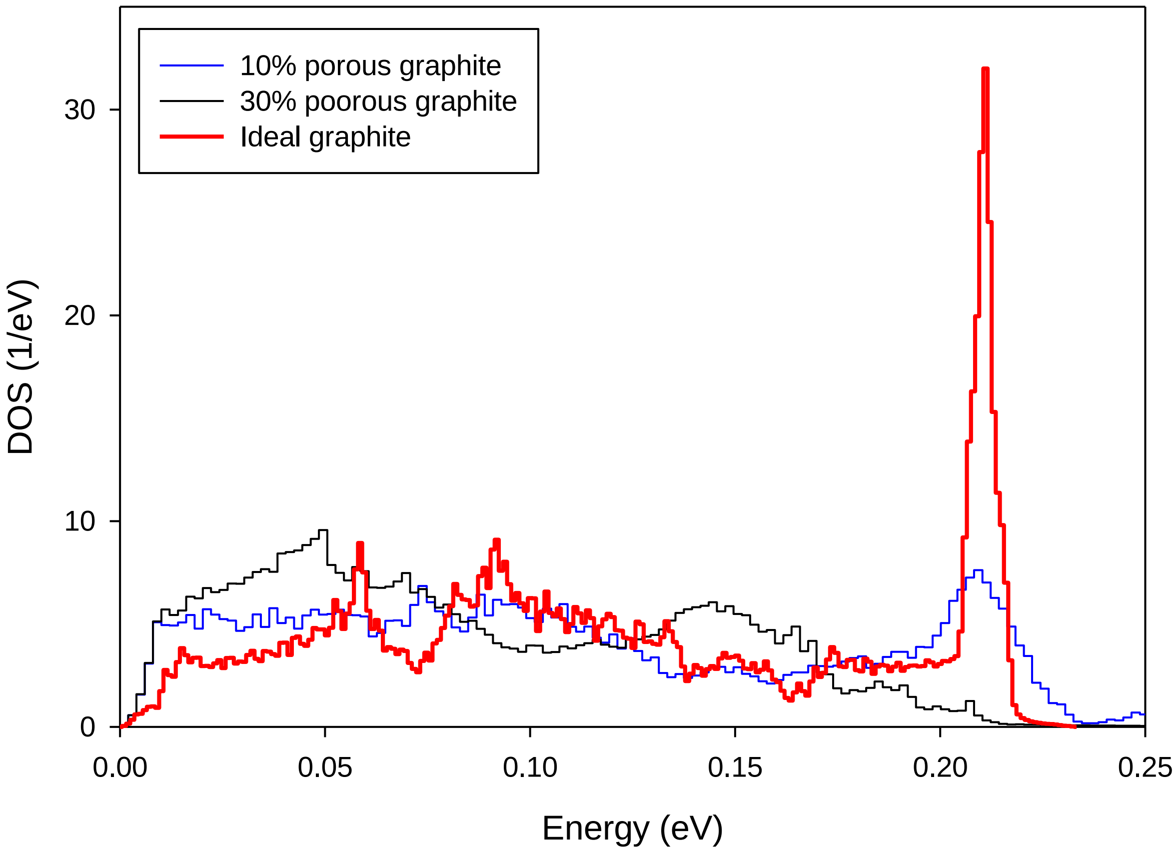

MD Simulation
$\{\vec{R}_j(t)\}$
$\{\vec{v}_j(t)\}$
Particle density autocorrelation
Velocity autocorrelation
$I(\vec{\kappa},t)$
Time Transform
Time Transform
$\rho(\omega)$
$S(\vec{\kappa},\omega)$
$S_s(\vec{\kappa},\omega)$

Density of States G(E)



Molecular Dynamics Models



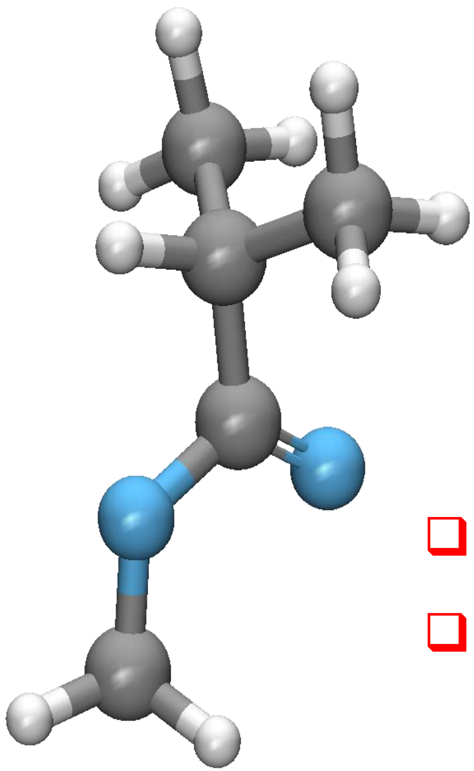


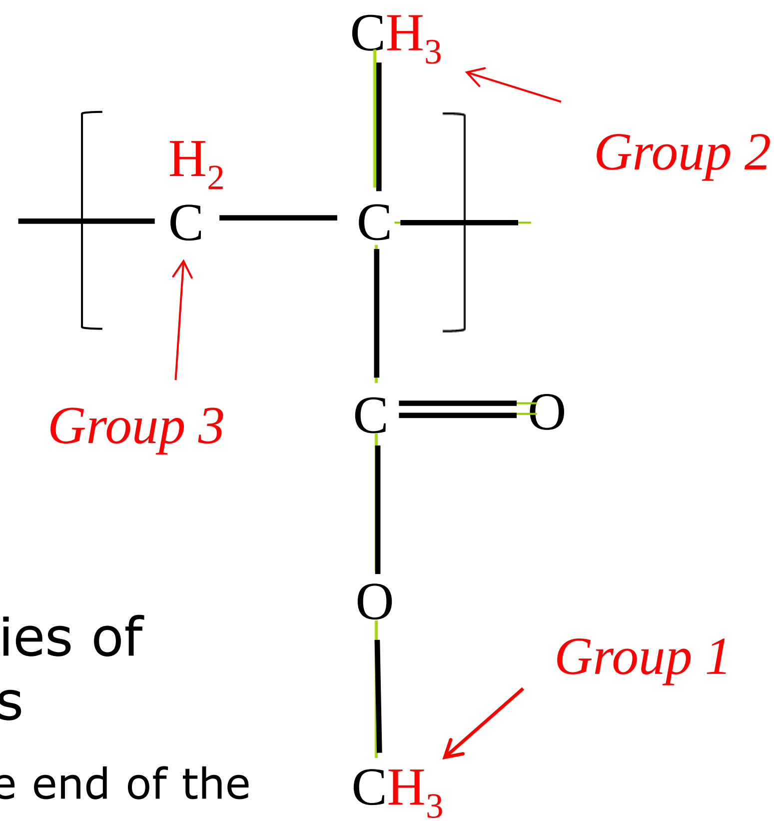
Materials Studied at NCSU

- Graphite, Beryllium (improvement on ENDF/B-VII)
  - Treatment of nuclear graphite (porous system)
  - Including coherent inelastic
- Silicon dioxide (New, contributed to NNDC/ENDF)
  - Support criticality safety analysis
- Silicon carbide (New, contributed to NNDC/ENDF)
  - Support advanced fuel cycle applications (e.g., FCM fuels)
- Thorium hydride, uranium-zirconium hydride, calcium hydride (New)
- Sapphire and bismuth (New)
  - Thermal neutron filters
- Solid methane (predictive analysis)
  - Cold neutron moderator
  - Captured phase I to II transformation upon cooling below 22 K

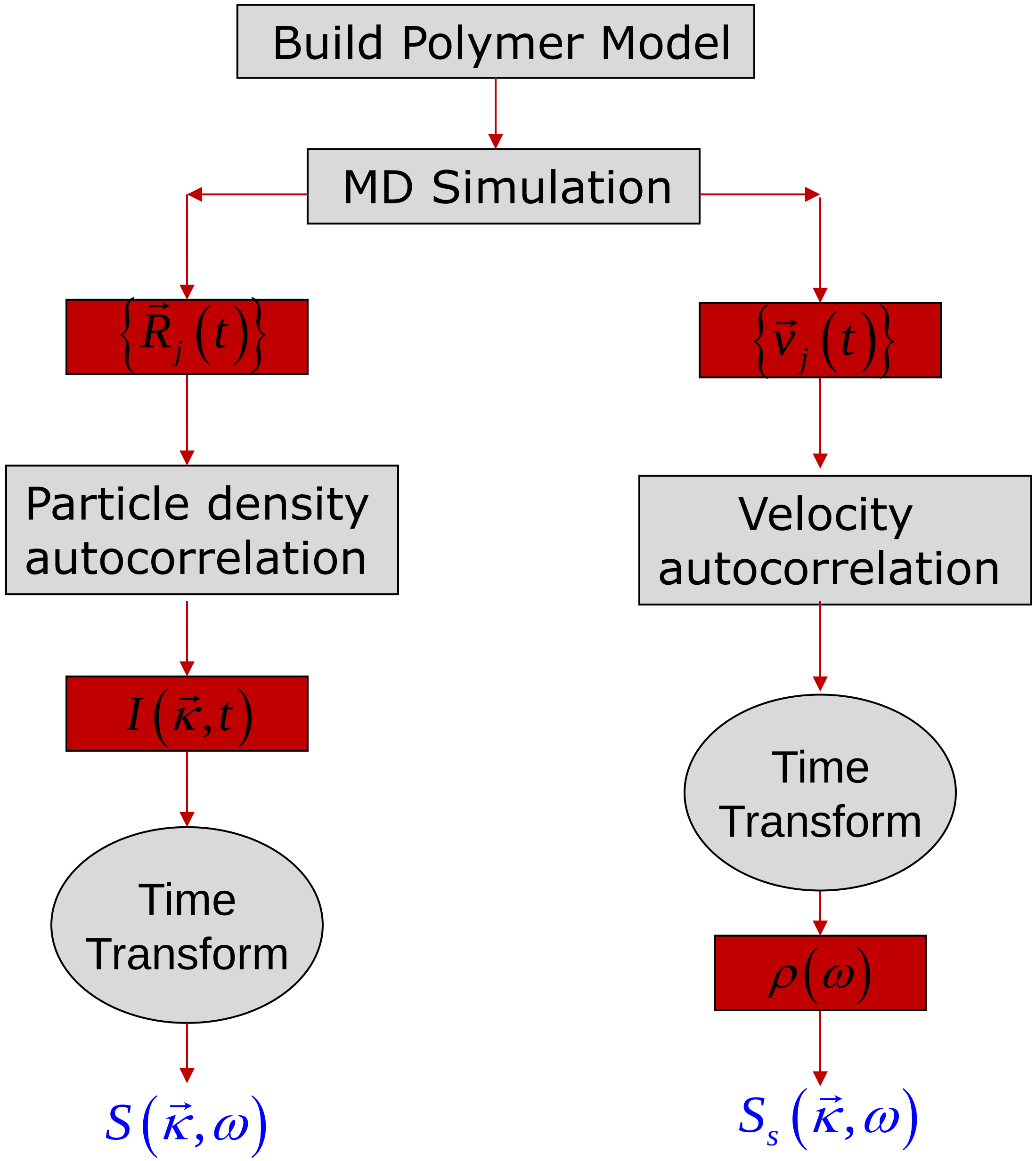
Lucite

Methyl Methacrylate Monomer



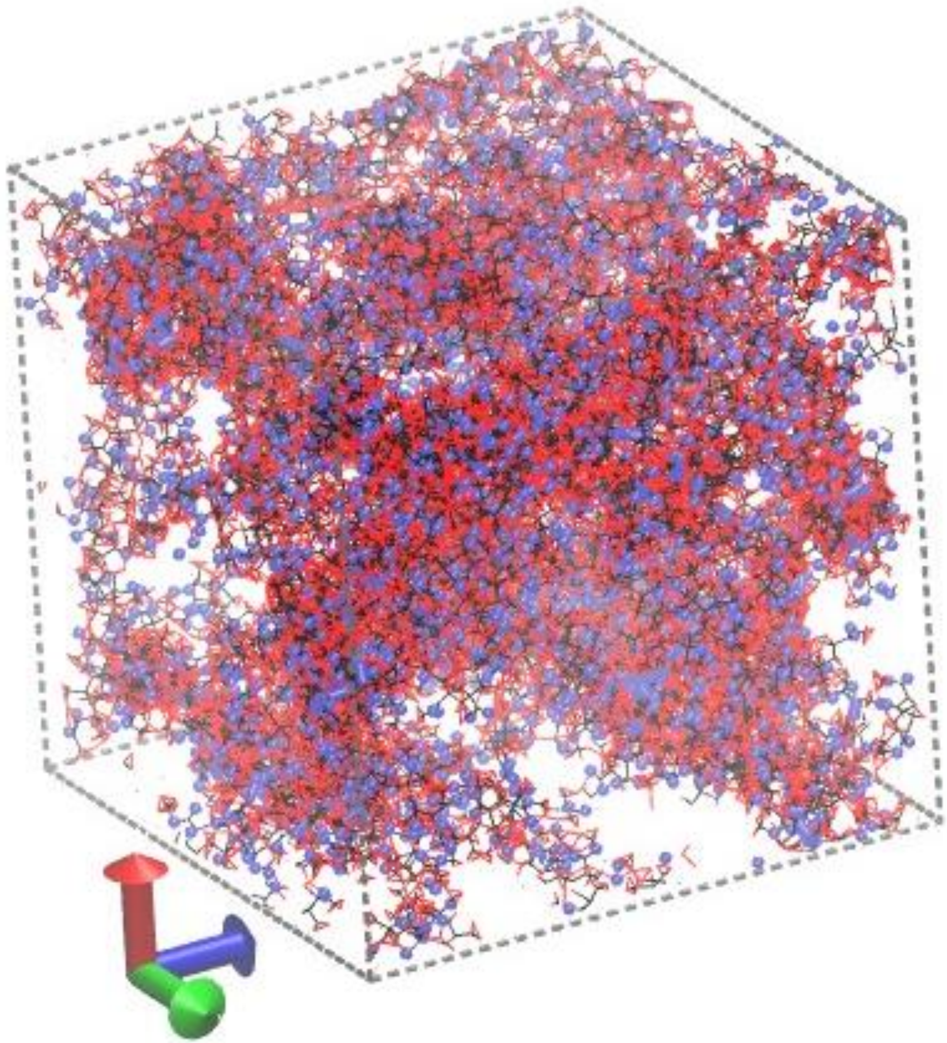


- $C5O2H8$
- 3 different species of Hydrogen atoms
  - Group 1: At the end of the longest chain
  - Group 2: At the end of the shorter chain
  - Group 3: On the backbone


Build Polymer Model
MD Simulation
$\left\{\vec{R}_j(t)\right\}$
$\left\{\vec{v}_j(t)\right\}$
Particle density autocorrelation
Velocity autocorrelation
$I(\vec{\kappa},t)$
Time Transform
Time Transform
$\rho(\omega)$
$S(\vec{\kappa},\omega)$
$S_s(\vec{\kappa},\omega)$

Lucite MD Model

- Amorphous poly(methyl methacrylate) polymer
- Molecular weight, minimum, 37,000 Mw (370 monomers long)
- 5 polymer chains



MD Potential Function

$$E = E_{vdw} + E_Q + E_B + E_A + E_T$$

$$E_{vdw} = AR^{-12} - BR^{-6}$$

$$E_Q = CQ_iQ_j/\epsilon R_{ij}$$

$$E_B = {}^1/_2\, k_0(R - R_0)^2$$

$$E_A = {}^1/_2\, K_{IJK}\left(cos\theta_{IJK} - cos\theta\right)^2$$

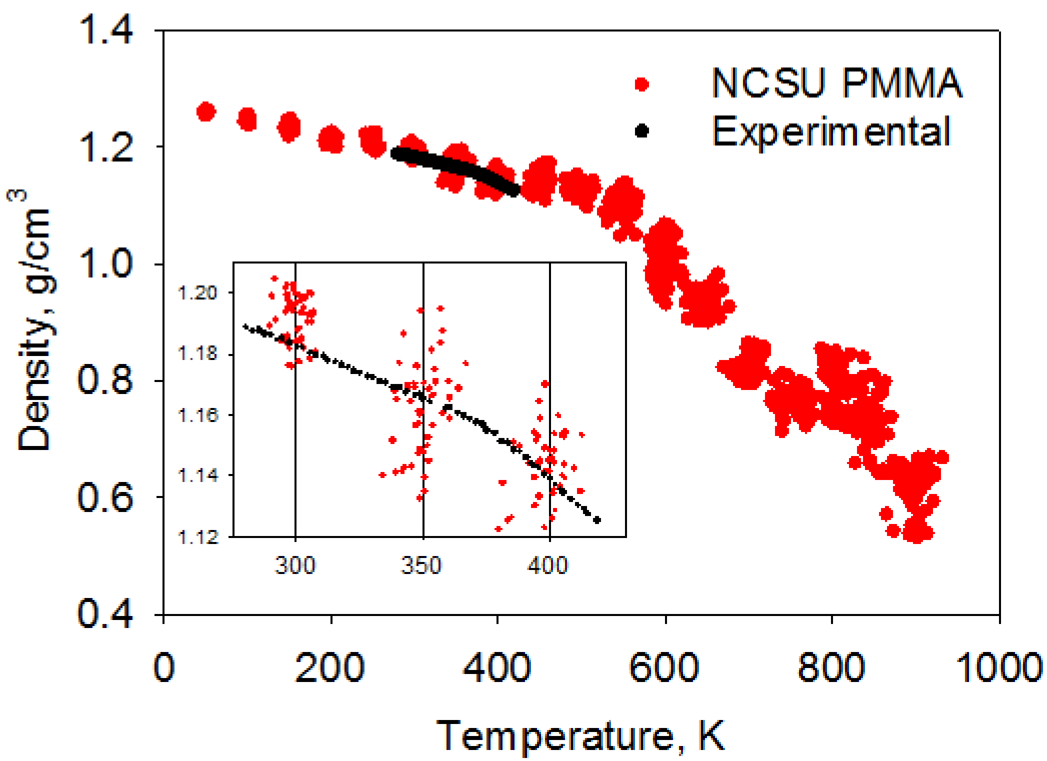
$$E_T = E_{IJKL} = {}^1/_2\, V_{JK}\{1 - cos[n_{JK}(\varphi - \varphi^0_{JK})]\}$$

Dreiding force field

L-J (VDW) term:

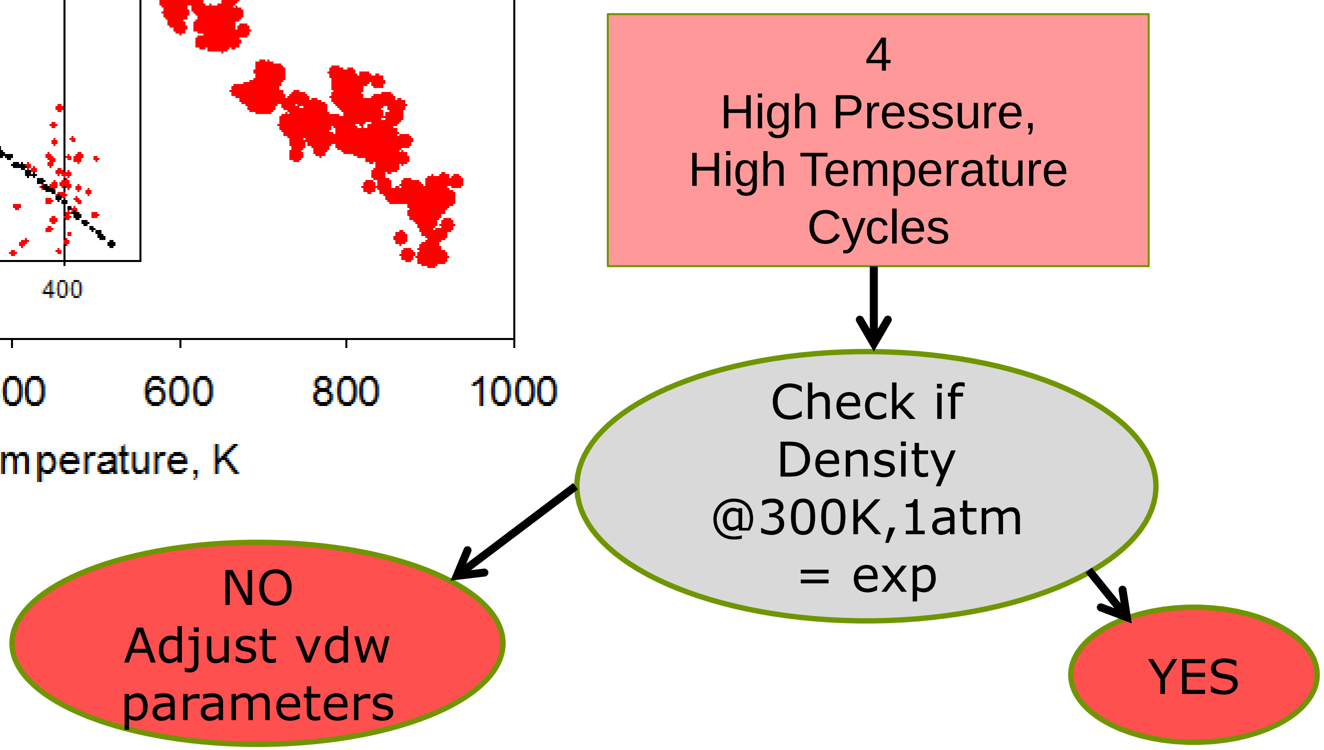
Fitted for the system to match the experimental density

Potential Parameterization



$T_g^{exp} \approx 400$ K

$T_g^{NCSU} = 400 - 500$ K



Computational Approach

$$C(t) = \left\langle \overrightarrow{v_j(0)} \cdot \overrightarrow{v_j(t)} \right\rangle = \frac{1}{N} \sum_{j-1}^{N} \overrightarrow{v_j(0)} \cdot \overrightarrow{v_j(t)}$$

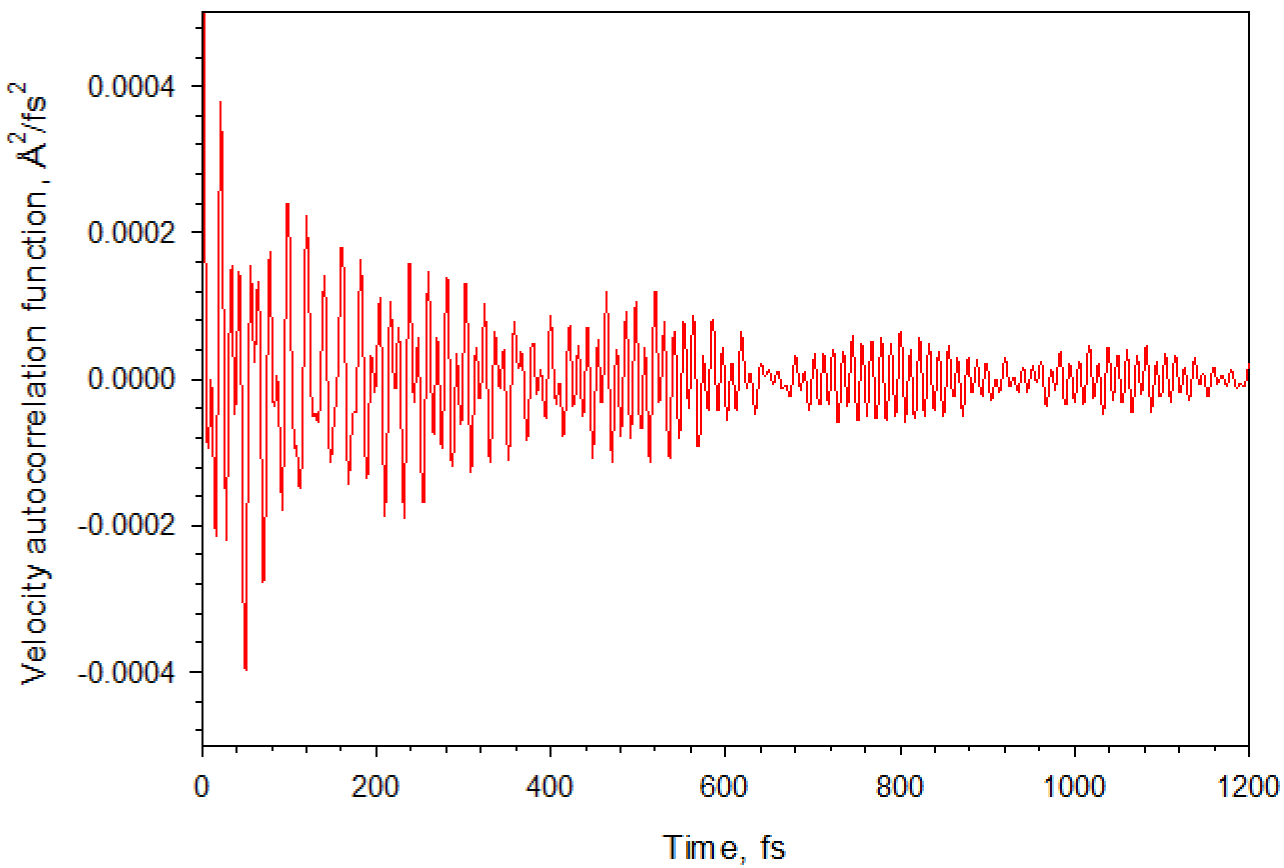
$$\rho(\omega) = \frac{M}{3\pi k_\beta T} \int_{-\infty}^{\infty} C(t) e^{-i\omega t} dt$$

Use NJOY/LEAPR

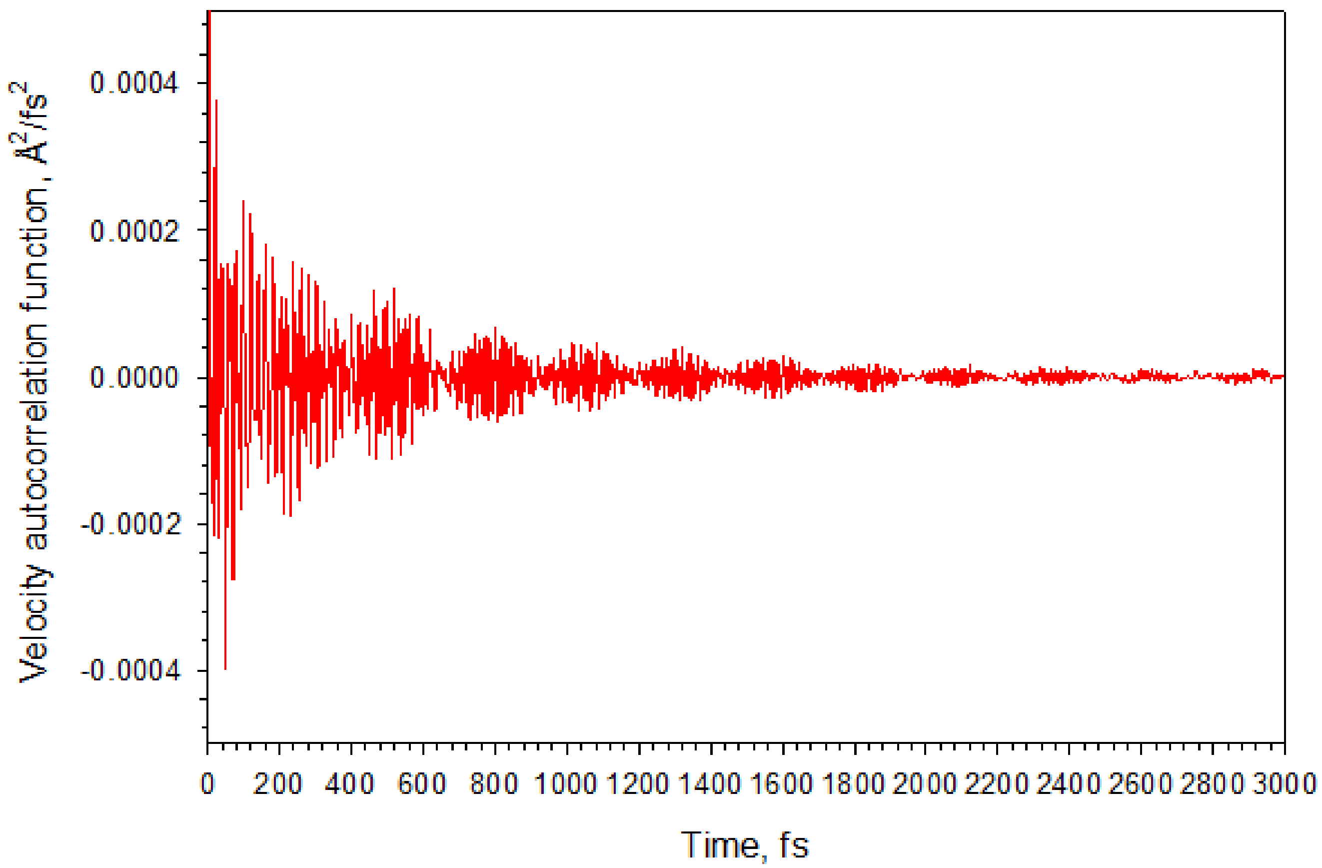
$$S(\alpha, \beta) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{-i\beta t} e^{-\gamma(t, \rho(\omega))} dt$$

$$\left. \frac{d^2\sigma}{d\Omega dE} \right|_{\text{inelastic}} = \frac{\sigma}{2k_B T} \sqrt{\frac{E'}{E}} e^{-\frac{\beta}{2}} S(\alpha, \beta)$$

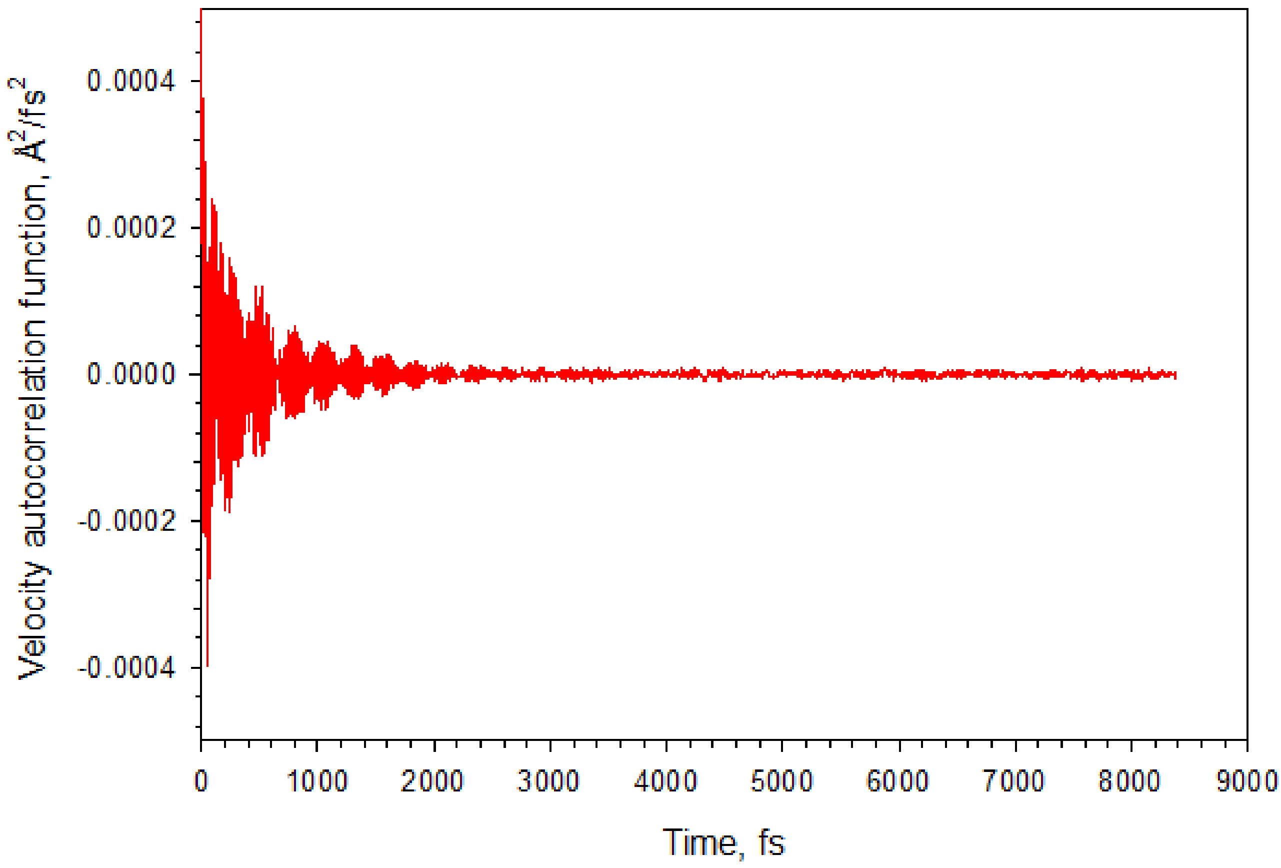
Lucite VACF


Velocity autocorrelation function, Å^2/fs^2
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
200
400
600
800
1000
1200
Time, fs

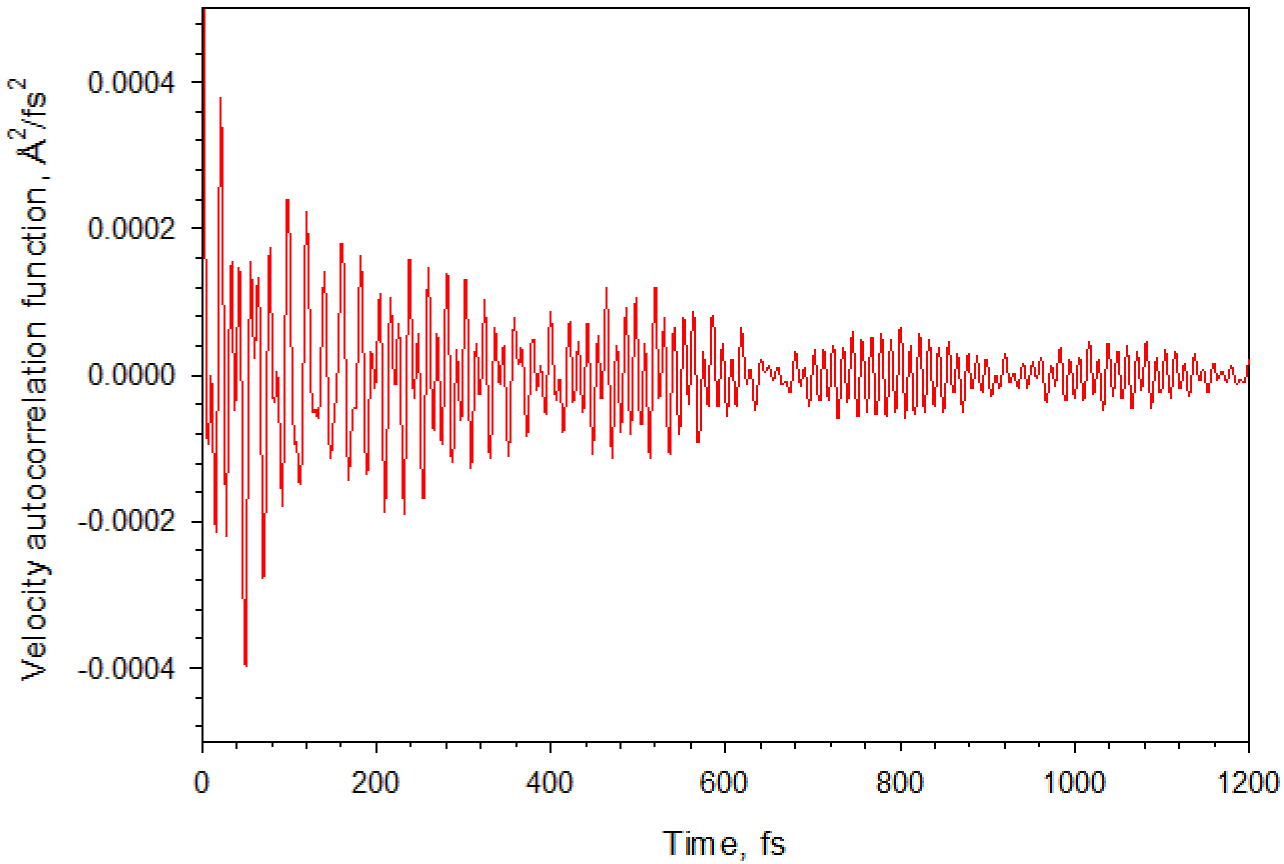
Lucite VACF


Velocity autocorrelation function, Å²/fs²
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
200
400
600
800
1000
1200
1400
1600
1800
2000
2200
2400
2600
2800
3000
Time, fs

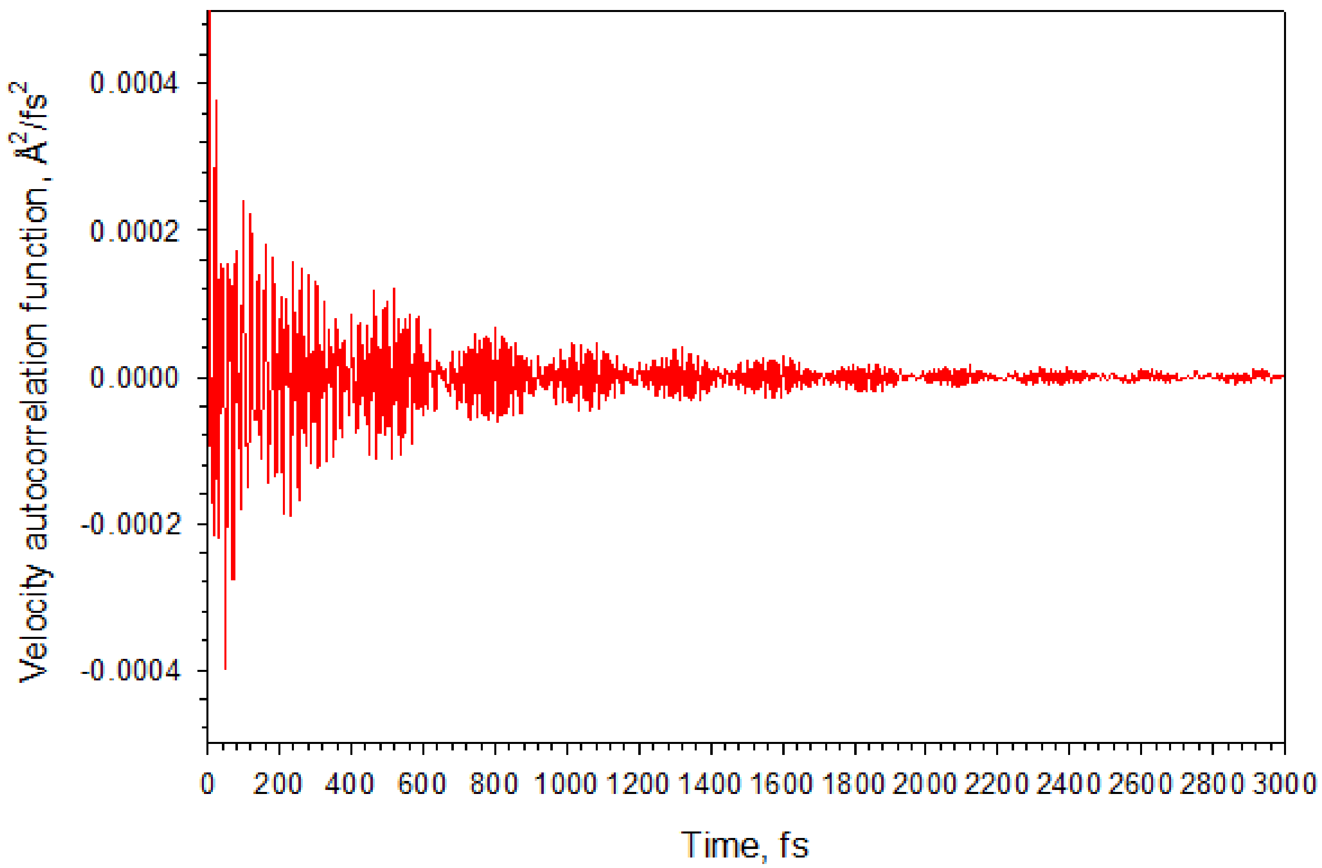
Lucite VACF


Velocity autocorrelation function, Å²/fs²
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
1000
2000
3000
4000
5000
6000
7000
8000
9000
Time, fs

Lucite VACF

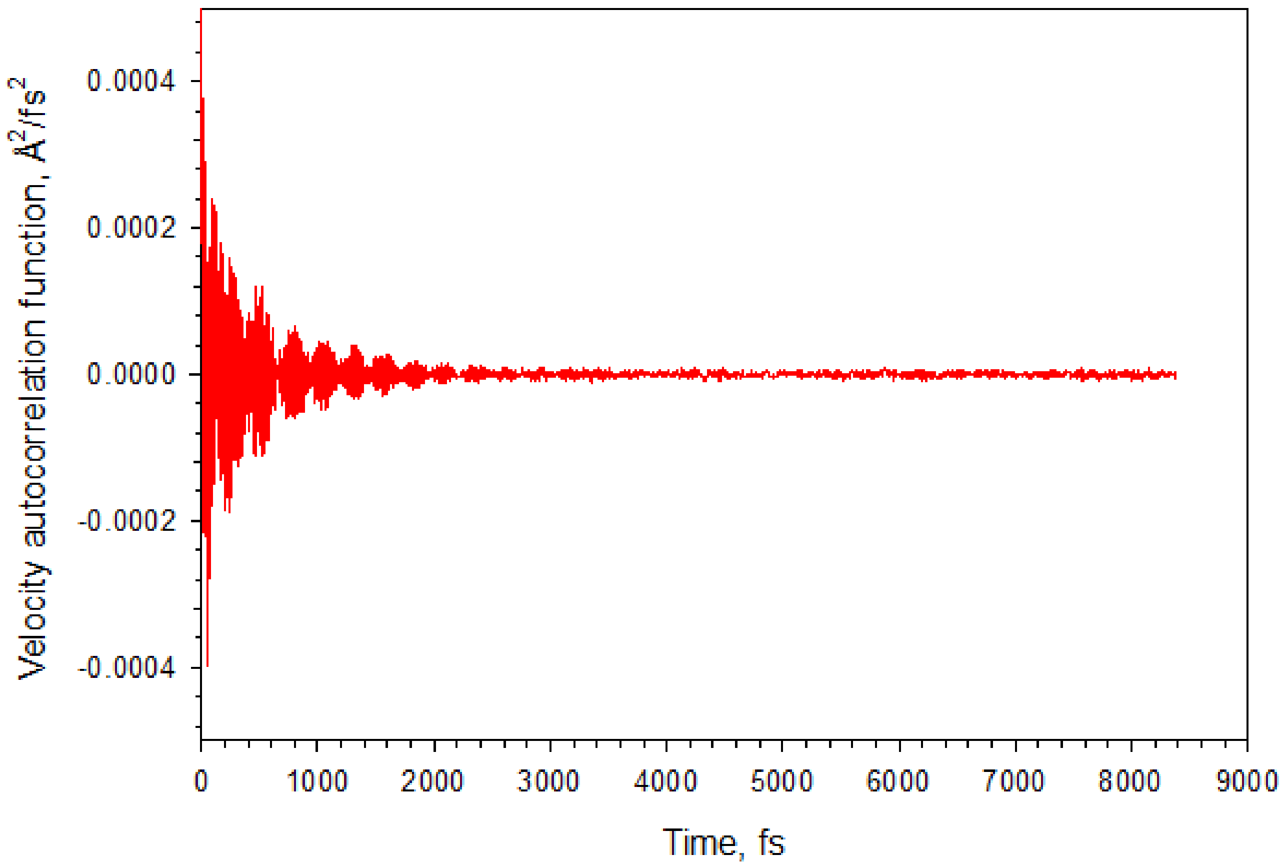

0.0004
0.0002
0.0000
-0.0002
-0.0004
Velocity autocorrelation function, Å^2/fs^2
0
200
400
600
800
1000
1200
Time, fs

Lucite VACF

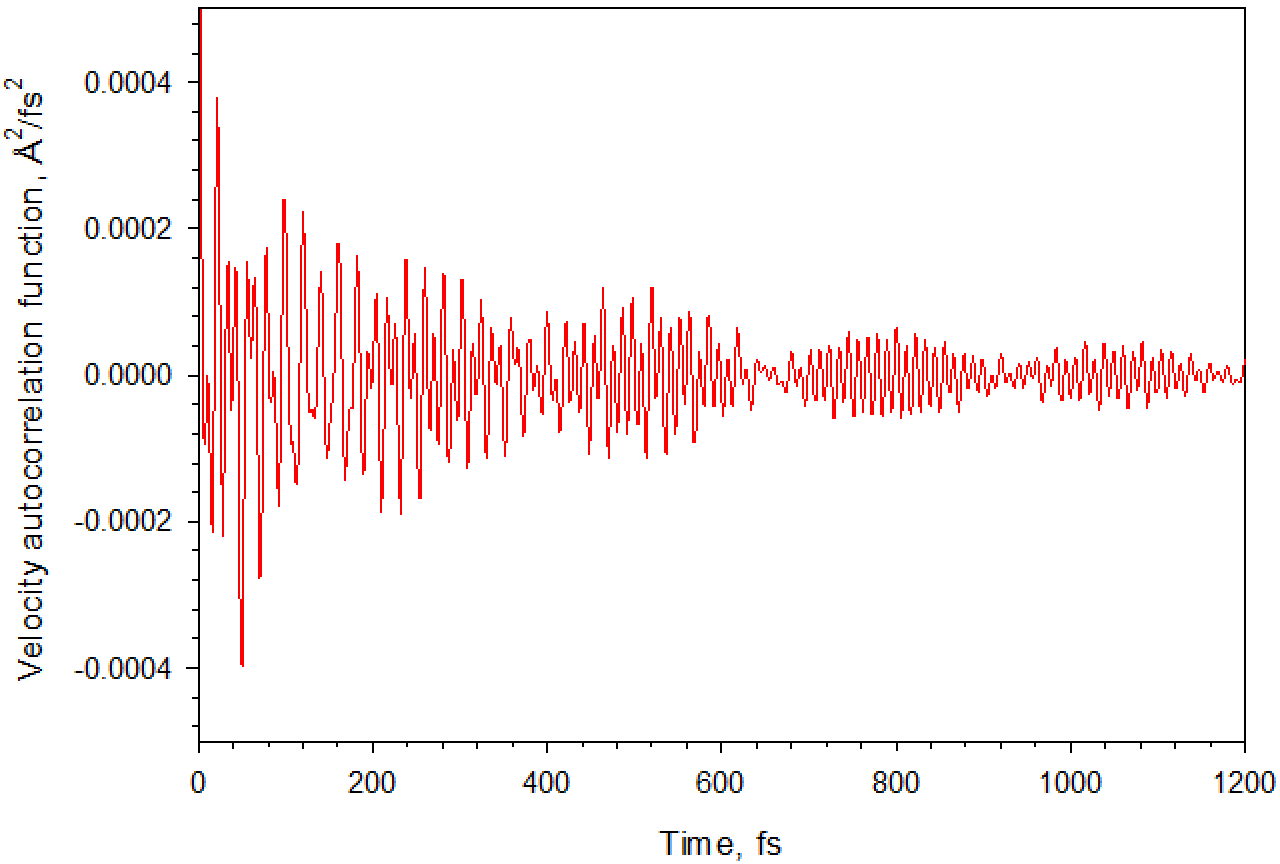


Velocity autocorrelation function, Å^2/fs^2
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
200
400
600
800
1000
1200
1400
1600
1800
2000
2200
2400
2600
2800
3000
Time, fs

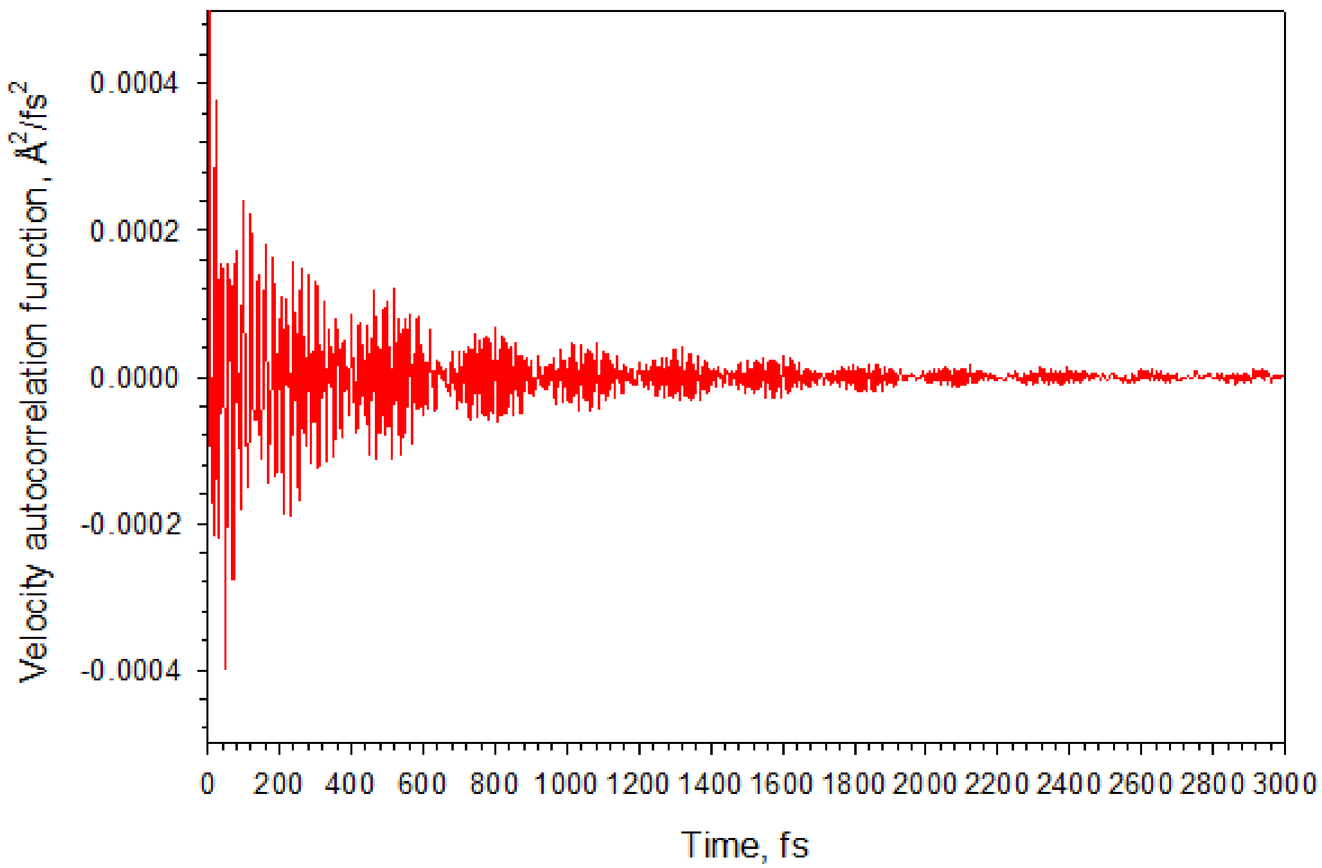
Lucite VACF


Velocity autocorrelation function, Å²/fs²
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
1000
2000
3000
4000
5000
6000
7000
8000
9000
Time, fs

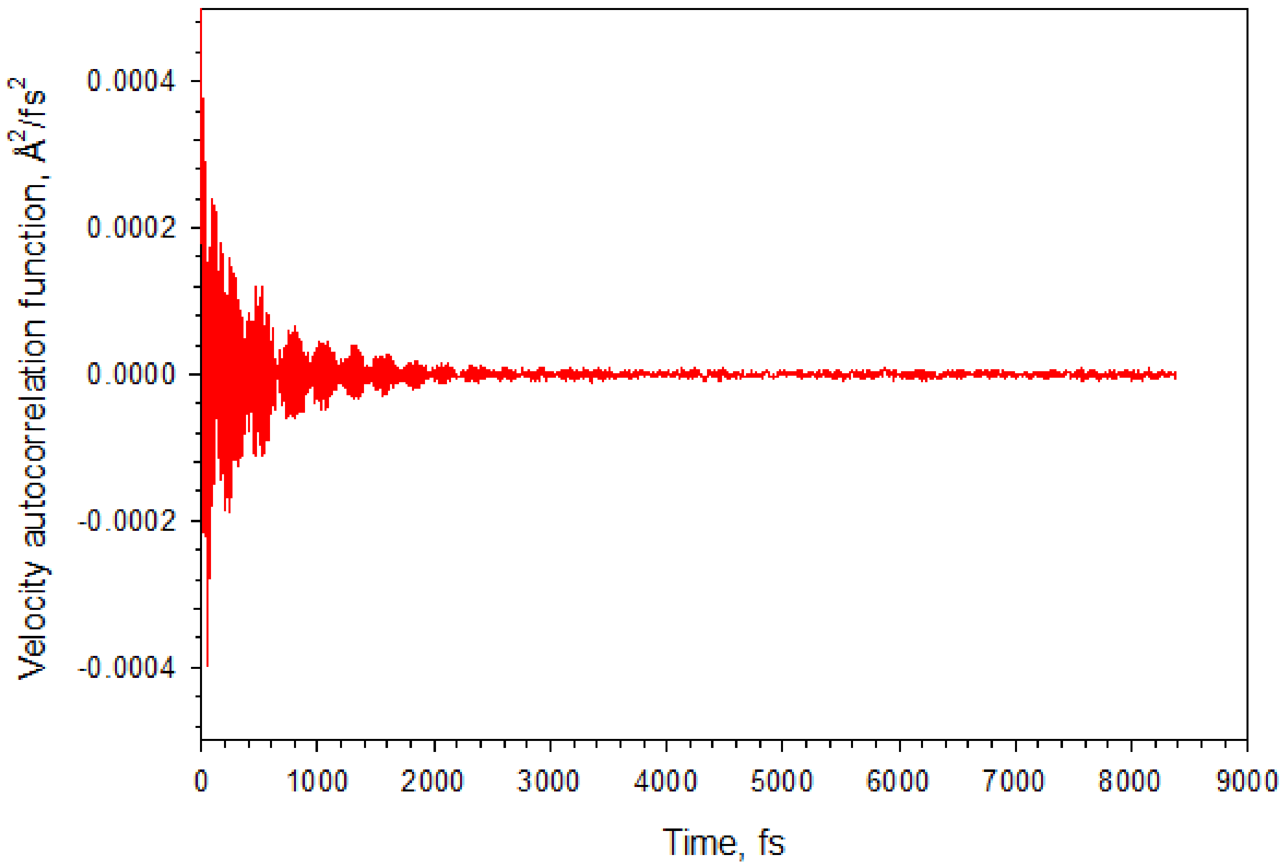
Lucite VACF


0.0004
0.0002
0.0000
-0.0002
-0.0004
Velocity autocorrelation function, Å²/fs²
0
200
400
600
800
1000
1200
Time, fs

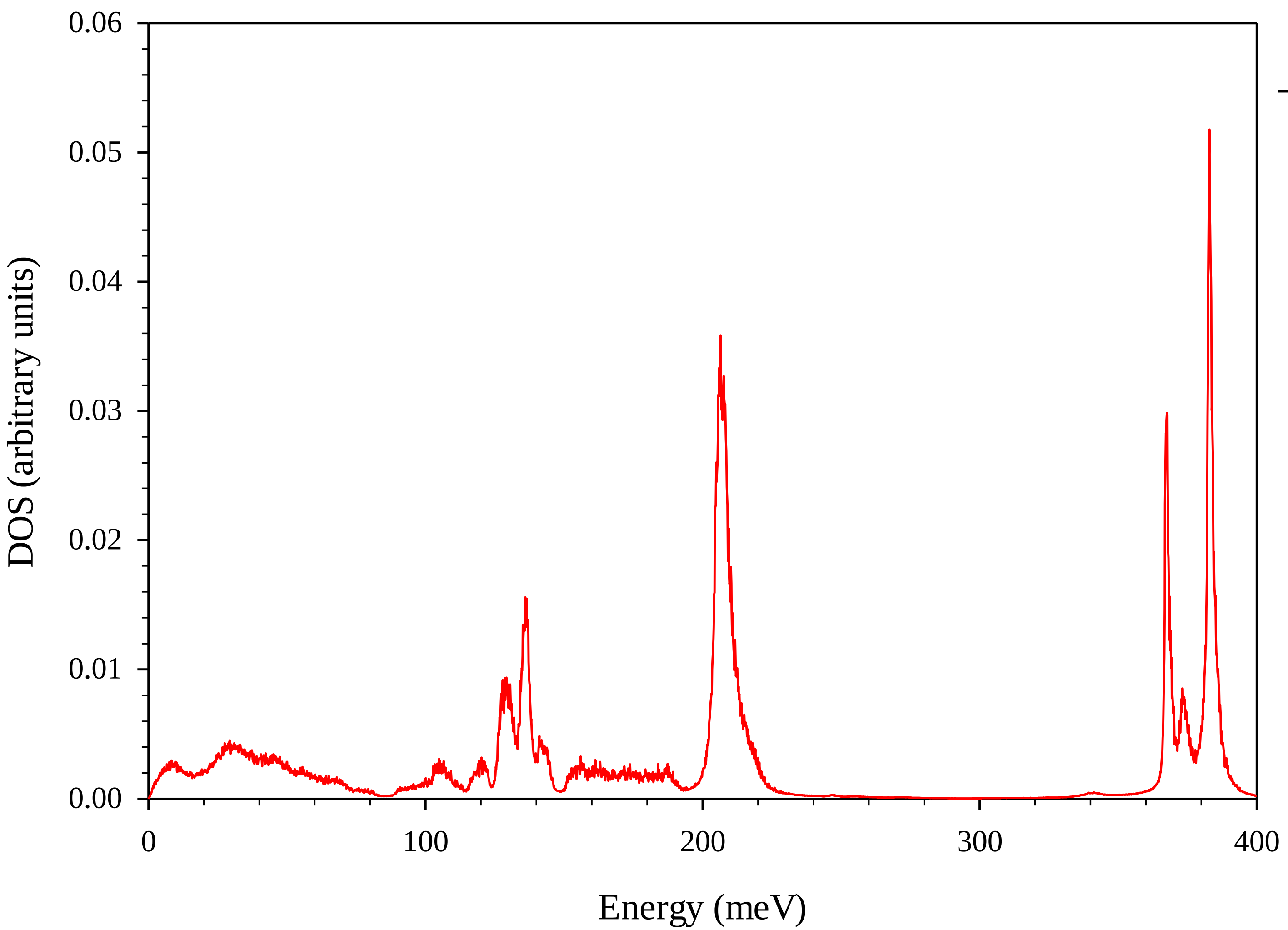
Lucite VACF

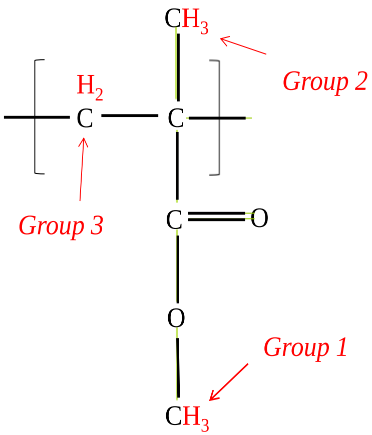

Velocity autocorrelation function, Å²/fs²
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
200
400
600
800
1000
1200
1400
1600
1800
2000
2200
2400
2600
2800
3000
Time, fs

Lucite VACF

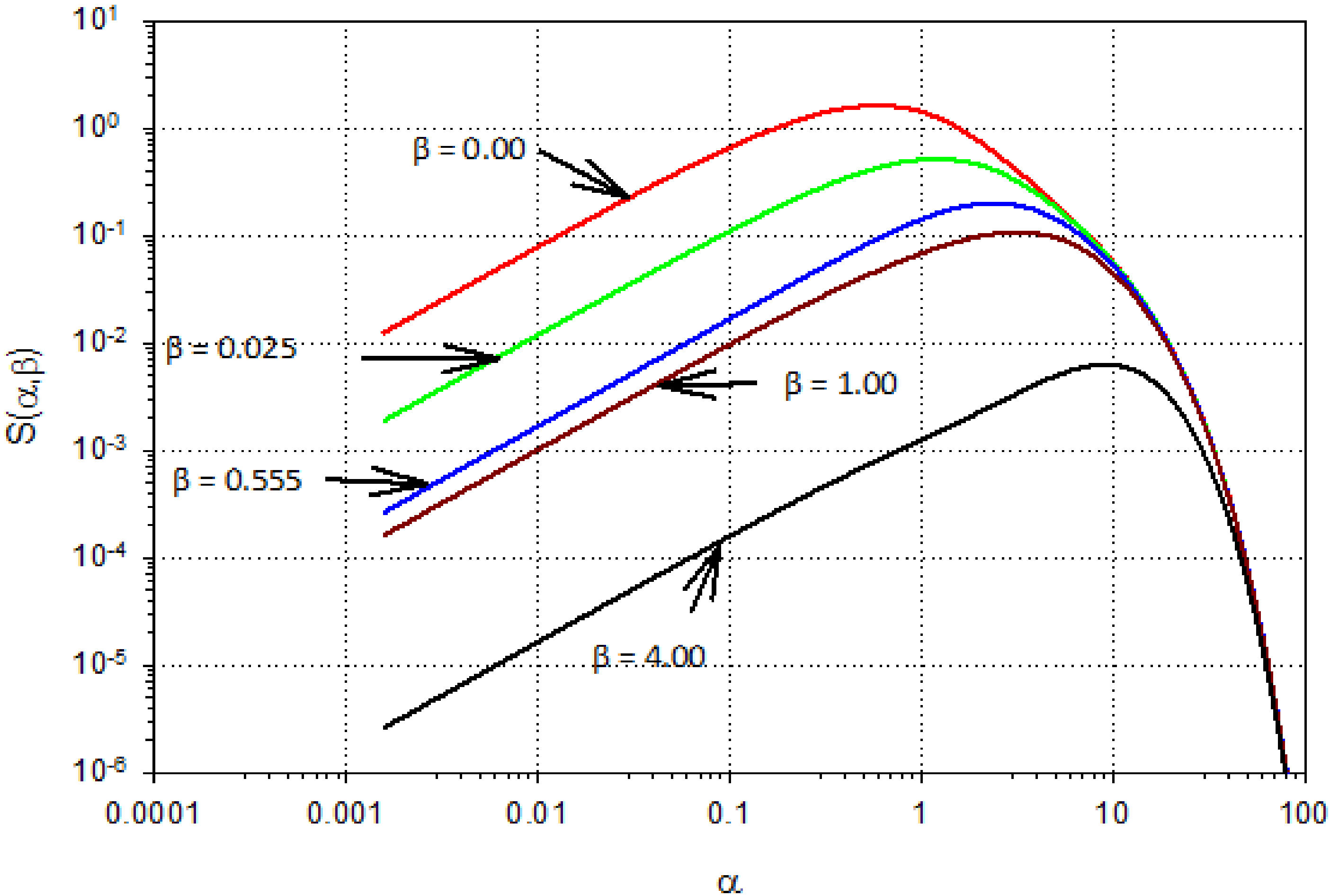

Velocity autocorrelation function, Å^2/fs^2
0.0004
0.0002
0.0000
-0.0002
-0.0004
0
1000
2000
3000
4000
5000
6000
7000
8000
9000
Time, fs

Density of States

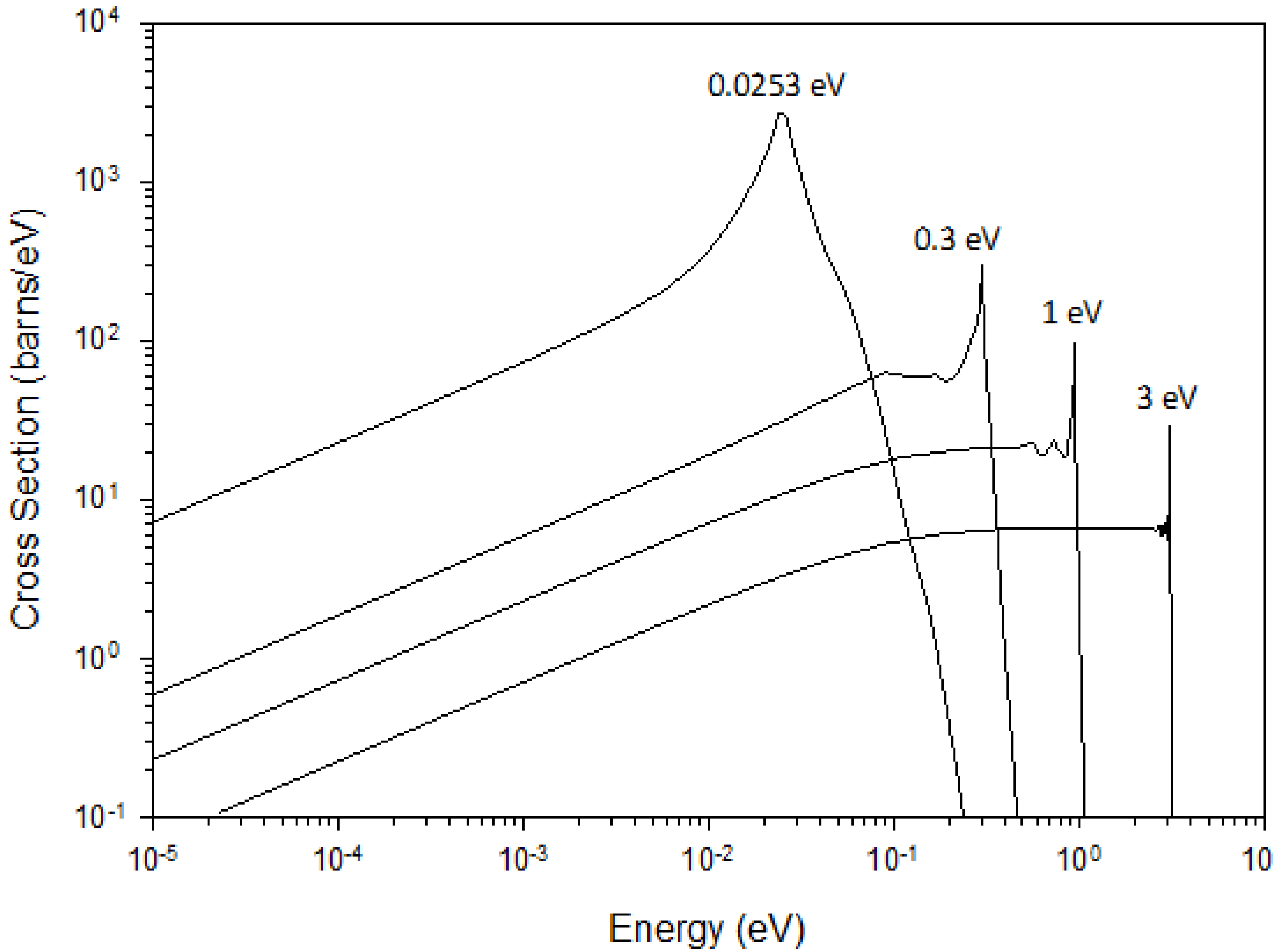




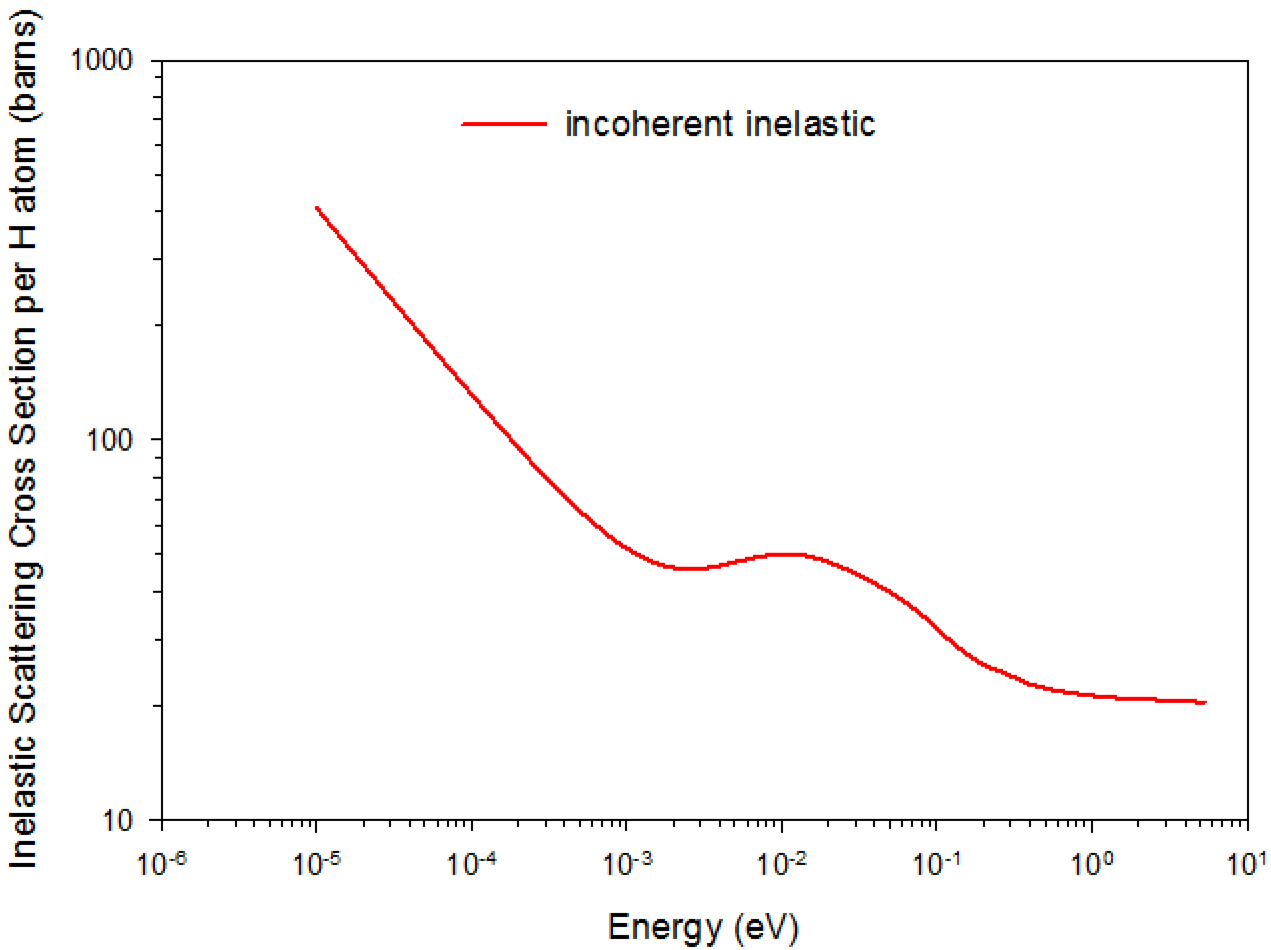
S(α,β) – Scattering Law



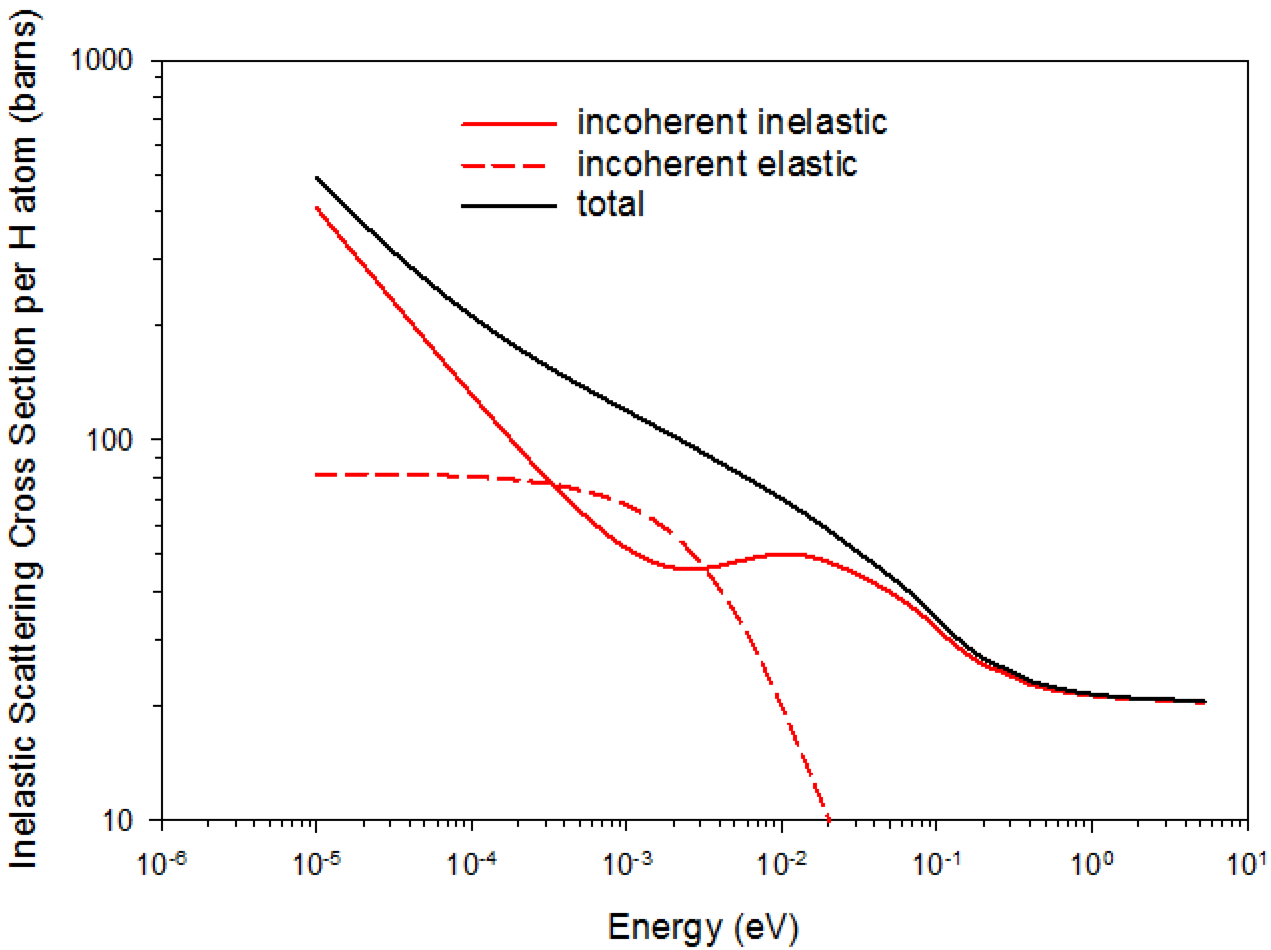
Differential Cross Section



Total Inelastic Scattering Cross Section



Total Scattering Cross Section



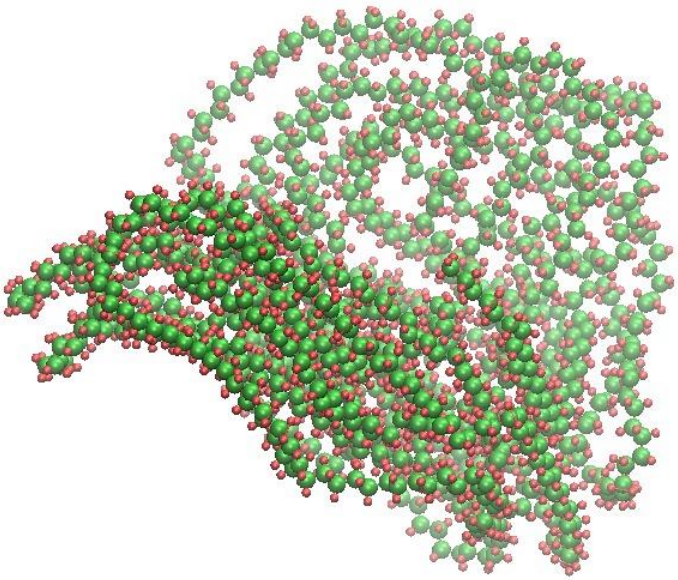
ACE Library

```
  Lu.00t    0.999167  2.5507E-08   02/20/15
xxxx                                                                    mat 125
   1001          0.        0          0.       0          0.       0          0.
      0          0.        0          0.       0          0.       0          0.
      0          0.        0          0.       0          0.       0          0.
      0          0.        0          0.       0          0.       0          0.
  30108          3        15         16          4        -1        1          0
      0          0         0          0          0         0        0          0
      1        111       220      29868      29989         0        0          0
      0          0         0          0          0         0        0          0
      0          0         0          0          0         0        0          0
      0          0         0          0          0         0        0          0
                 109  1.00000000000E-11  1.78000000000E-11  2.50000000000E-11
   3.50000000000E-11  5.00000000000E-11  7.00000000000E-11  1.00000000000E-10
   1.26000000000E-10  1.60000000000E-10  2.00000000000E-10  2.53000000000E-10
   2.97000000000E-10  3.50000000000E-10  4.20000000000E-10  5.06000000000E-10
   6.15000000000E-10  7.50000000000E-10  8.70000000000E-10  1.01200000000E-09
   1.23000000000E-09  1.50000000000E-09  1.80000000000E-09  2.03000000000E-09
   2.27700000000E-09  2.60000000000E-09  3.00000000000E-09  3.50000000000E-09
   4.04800000000E-09  4.50000000000E-09  5.00000000000E-09  5.60000000000E-09
   6.32500000000E-09  7.20000000000E-09  8.10000000000E-09  9.10800000000E-09
   1.00000000000E-08  1.06300000000E-08  1.15000000000E-08  1.23970000000E-08
   1.33000000000E-08  1.41700000000E-08  1.50000000000E-08  1.61920000000E-08
   1.82000000000E-08  1.99000000000E-08  2.04930000000E-08  2.15000000000E-08
   2.28000000000E-08  2.53000000000E-08  2.80000000000E-08  3.06130000000E-08
   3.38000000000E-08  3.65000000000E-08  3.95000000000E-08  4.27570000000E-08
   4.65000000000E-08  5.00000000000E-08  5.69250000000E-08  6.25000000000E-08
   6.90000000000E-08  7.50000000000E-08  8.19720000000E-08  9.00000000000E-08
   9.60000000000E-08  1.03500000000E-07  1.11573000000E-07  1.20000000000E-07
   1.28000000000E-07  1.35500000000E-07  1.45728000000E-07  1.60000000000E-07
   1.72000000000E-07  1.84437000000E-07  2.00000000000E-07  2.27700000000E-07
   2.51039000000E-07  2.70530000000E-07  2.90750000000E-07  3.01133000000E-07
   3.20642000000E-07  3.57681000000E-07  3.90000000000E-07  4.17035000000E-07
   4.50000000000E-07  5.03258000000E-07  5.60000000000E-07  6.25000000000E-07
   7.00000000000E-07  7.80000000000E-07  8.60000000000E-07  9.50000000000E-07
   1.05000000000E-06  1.16000000000E-06  1.28000000000E-06  1.42000000000E-06
   1.55000000000E-06  1.70000000000E-06  1.85500000000E-06  2.02000000000E-06
   2.18000000000E-06  2.36000000000E-06  2.59000000000E-06  2.85500000000E-06
   3.12000000000E-06  3.42000000000E-06  3.75000000000E-06  4.07000000000E-06
   4.46000000000E-06  4.90000000000E-06  4.03040400000E+02  3.02924116709E+02
   2.55846264285E+02  2.16793828099E+02  1.82069700000E+02  1.54667743636E+02
   1.30374300000E+02  1.16916204366E+02  1.04665636038E+02  9.45428600000E+01
   8.52140318505E+01  7.95217413784E+01  7.42313428635E+01  6.89817840000E+01
   6.42258331097E+01  5.98817859922E+01  5.60964431935E+01  5.36921384618E+01
   5.15350046384E+01  4.93079173888E+01  4.76385095017E+01  4.66213927786E+01
   4.62110047309E+01  4.60018068570E+01  4.59769073341E+01  4.61676914453E+01
   4.66030757828E+01  4.71728973600E+01  4.76619800000E+01  4.81835200000E+01
   4.87515445214E+01  4.93353002010E+01  4.98805999380E+01  5.02818346155E+01
   5.05706456665E+01  5.06890400000E+01  5.07052035865E+01  5.06625410857E+01
   5.05534983937E+01  5.03787705162E+01  5.01694716065E+01  4.99406058832E+01
```

Polyethylene

Polyethylene MD Model

- Partially amorphous, partly crystalline structure, made up of ethylene monomers, C_2H_4
- Polyethylene is the most common plastic, ubiquitously used for packaging, bags and bottles
- Molecular weight, 10,000-100,000 Mw (at least 350 monomers long)



MD Potential Function

$$E = E_{vdw} + E_Q + E_B + E_A + E_T$$

$$E_{vdw} = AR^{-12} - BR^{-6}$$

$$E_Q = CQ_iQ_j/\epsilon R_{ij}$$

$$E_B = {}^{1}/_{2}\, k_0(R - R_0)^2$$

$$E_A = {}^{1}/_{2}\, K_{IJK}\left(cos\theta_{IJK} - cos\theta\right)^2$$

$$E_T = E_{IJKL} = {}^{1}/_{2}\, V_{JK}\{1 - cos\left[n_{JK}\left(\varphi - \varphi_{Jk}^{0}\right)\right]\}$$

Dihedral barrier term:

Fitted for the system to match the experimental amorphous/crystal composition

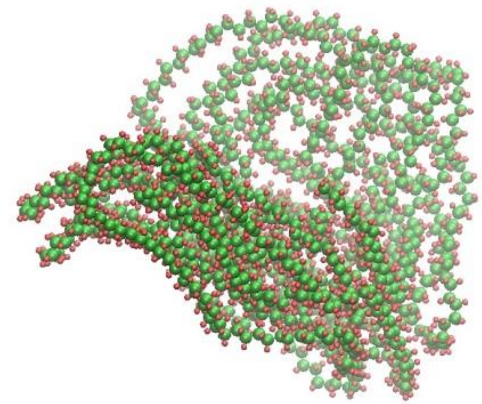
Parameterization

Dihedral barrier of 2 kcals/mole

↓

~ 50% crystallinity

→

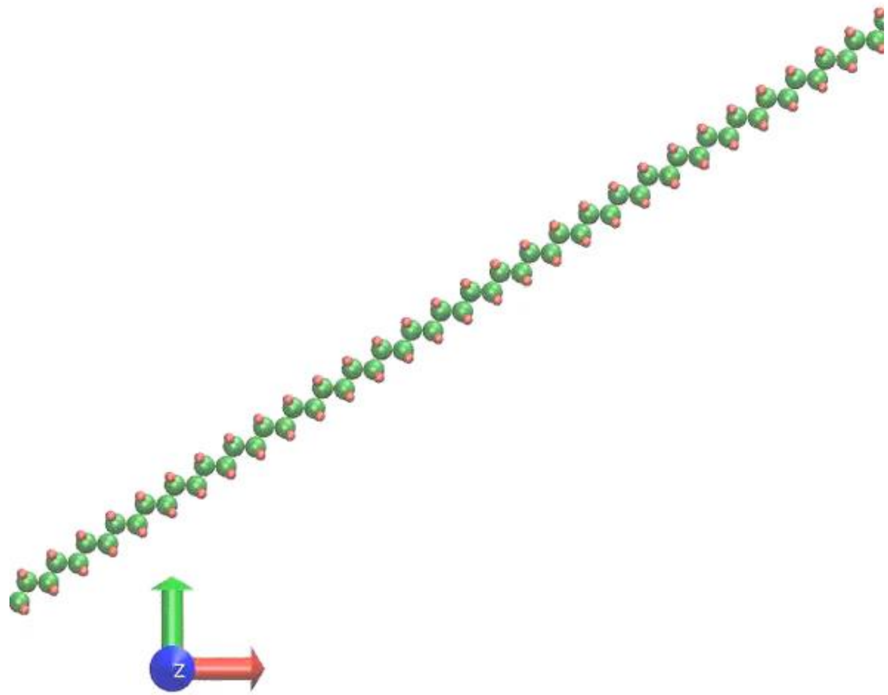


Dihedral barrier of 3 kcals/mole

↓

~100% crystallinity

→


z

Summary

- Developed a modern approach for thermal neutron cross section calculations based on the use of atomistic simulations
  - Ab initio quantum mechanics
  - Molecular dynamics
- The approach is predictive
  - New materials
  - All states of matter (solid, liquid, gas)
  - Imperfect structure
- Evaluating NCSP materials
  - Silicon dioxide completed
  - Lucite completed
  - Polyethylene initiated

Future

- Continue to support the data needs of NCSP and the nuclear science and engineering community
- Develop the “next generation” platform for thermal neutron scattering analysis
  - Free from approximations
  - Flexible to use various types of input
  - Able to seamlessly integrate with neutron transport tools